



Dissolved organic matter removal during coal slag additive soil aquifer treatment for secondary effluent recharging: Contribution of aerobic biodegradation



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ABSTRACT

Recycling wastewater treatment plant (WWTP) effluent at low cost via the soil aquifer treatment (SAT), which has been considered as a renewable approach in regenerating potable and non-potable water, is welcome in arid and semi-arid regions throughout the world. In this study, the effect of a coal slag additive on the bulk removal of the dissolved organic matter (DOM) in WWTP effluent during SAT operation was explored via the matrix configurations of both coal slag layer and natural soil layer. Azide inhibition and XAD-resins fractionation experiments indicated that the appropriate configuration designing of an upper soil layer (25 cm) and a mixture of soil/coal slag underneath would enhance the removal efficiency of adsorption and anaerobic biodegradation to the same level as that of aerobic biodegradation (31.7% vs 32.2%), while it was only 29.4% compared with the aerobic biodegradation during traditional 50 cm soil column operation. The added coal slag would preferentially adsorb the hydrophobic DOM, and those adsorbed organics could be partially biodegraded by the biomass within the SAT systems. Compared with the relatively lower dissolved organic carbon (DOC), ultraviolet light adsorption at 254 nm (UV-254) and trihalomethane formation potential (THMFP) removal rate of the original soil column (42.0%, 32.9%, and 28.0%, respectively), SSL2 and SSL4 columns would enhance the bulk removal efficiency to more than 60%. Moreover, a coal slag additive in the SAT columns could decline the aromatic components (fulvic-like organics and tryptophan-like proteins) significantly.

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1. Introduction

Water shortage and controlling wastewater pollution are two serious environmental issues in the arid and semi-arid regions (Bakopoulou et al., 2011; Dawadi and Ahmad, 2013). In addition, efficient reuse of secondary effluent from wastewater treatment plants (WWTP) is rapidly becoming a necessity for many municipalities (Wei et al., 2009a; Zhang et al., 2010). Because of its relatively easy operation, low construction/operational cost, and high efficiency, soil aquifer treatment (SAT) has recently been employed

to treat WWTP effluents via artificial water recharging, during which water quality is improved via the integrated functions of biochemical and physical processes (Nijhawan et al., 2013; Hubner et al., 2014). Reclaimed water from the SAT could be recycled for potable and non-potable uses after simple disinfection. Because there is strong evidence of the correlation between carcinogenic disinfection by-product (DBP) formation potential and the existence of dissolved organic matter (DOM) within the reclaimed water (Xue et al., 2008; Verlicchi et al., 2009), the search for new and innovative operation of SAT have focused attention on the environmental behavior and transformation of DOM.

DOM in the WWTP effluent is a heterogeneous mixture of organics, which consist of high-molecular-weight humic substances, polysaccharides, hydrophilic acids, aromatic proteins, and amino acids (Xue et al., 2008). DOM plays an important role in SAT

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reclaimed systems, such as metals mobilization, pollutant degradation, and carbon release and utilization; moreover, it was also the main precursor of DBPs during chlorination of the reclaimed water (Sobhani et al., 2012). Biodegradation and adsorption processes are both contributors to DOM reduction during SAT operation (Fox et al., 2005), with biodegradation being dominant during traditional SAT operation. Being aware of the above described limitations, enhanced adsorption with activated carbon has been widely used in SAT operations (Schreiber et al., 2005; Acero et al., 2010). Taking environmentally acceptable and economical elements into consideration, cheaper adsorbents, such as red mud, fly ash, carbon slurry, peat, coal slag, and activated rice husks, have been attempted as substitutes of activated carbon (Cha et al., 2006; Gupta and Suhas, 2009; Wei et al., 2010; Geyikci et al., 2012). Because of its adsorptive characteristics and local availability (annual production of coal slag in China is estimated at approximately 0.5 billion tons), coal slag has been recently recognized as an alternative to granular activated carbon (GAC) (Dimitrova, 2002).

Coal slag and its modified products exhibited adsorptive properties and have been widely used in various industrial and sewage wastewater treatments for replacing currently used GAC (Liu et al., 2010; Li et al., 2011), and adsorption of dye, nickel, phosphorus, and lead are examples of coal slag use (Ortiz et al., 2001). A recent study by Genc and Oguz (2010) revealed that the removal efficiency of the dye Acid Yellow 99 could reach 50% at a slag dosage of 10 g/L, and its adsorption characteristics could be modeled with the Langmuir isotherm. In addition, physical and chemical surface modifications could enhance the dyes' adsorption efficiency (Xue et al., 2009b). Several studies have also shown that the adsorption of phosphorus by blast furnace slag follows a pseudo-second-order kinetic model (Kostura et al., 2005). Recently, an active melter slag filter was established in New Zealand to treat the effluent from waste stabilization ponds (McDowell et al., 2007), and 54%–84% of the bulk phosphorus was adsorbed (Pratt et al., 2007). Liu et al. (2010) demonstrated that the removal efficiency of Pb^{2+} (100 mg/L) would reach 96% under 30 g/L slag dosage. Moreover, Cha et al. (2006) stated that the addition of a steel slag would sharply enhance the bulk removal of organics during SAT treatment. Although the use of coal slag as a low cost adsorbent is increasing, few reports have been published on the application of coal slag modified SAT for secondary effluent recharging. In order to clarify the biodegradation removal mechanism of DOM during SAT operation, ultraviolet light (UV) and excitation-emission matrix (EEM) have been traditionally used for elucidating the biodegradability of dissolved organic carbon (DOC) (Azwa et al., 2013). To the authors' knowledge, no research has been performed to quantify the contribution of biodegradation and adsorption in coal slag additive SAT, which is essential for further studying the removal mechanism of DOM during SAT operation (McDowell et al., 2006; Xue et al., 2008).

The goal of work described here was to assess the contribution of a coal slag additive to the removal of DOM related trihalomethane (THM) precursors during SAT operation. The effect of coal slag modification on biomass activity, as well as the contribution of biodegradation and adsorption in DOM removal was also analyzed. Fluorescence spectrum analysis was applied to characterize the chemical structural variation of DOM fractions.

2. Materials and methods

2.1. Experimental materials

Experimental coal slag was collected from the boiler house of the Harbin Institute of Technology (Harbin, China). Prior to and in preparation for its use in the experiments, the coal slag was screened

between 5 and 8 mm, washed and then dried at ambient temperature. The main composition of the coal slag was determined by X-ray fluorescence (XRF), resulting in a following chemical composition: 54.8–55.9% SiO_2 , 22.4–27.2% Al_2O_3 , 1.7–1.8% CaO, 0.5% MgO, 3.9–5.1% Fe_2O_3 , 1.8–2.0% K_2O , 0.6% Na_2O , and 1.1–1.2% TiO_2 (w/w), with a loss on ignition (LOI) of 4.7–8.6%. An X-ray diffraction (XRD) spectrum showed abundant quartz (SiO_2), mullite ($Al_6Si_2O_{13}$), sodium aluminum phosphate ($Na_4Al_2Si_2O_9$), iron oxide (Fe_2O_3), and aluminum oxide (Al_2O_3). The density and Brunauer–Emmett–Teller (BET) specific surface of this coal slag as determined by standard methods was found to be approximately 2560–3320 kg/m^3 and 2.3 m^2/g (particular size of 120–250 mesh), respectively. Scanning electron microscope (SEM) images indicated a rugged and porous surface characteristic of the experimental coal slag (see Fig. S1, Supplementary material), which was observed by a MX2600FE scanning electron microscope (Camscan Company, U.K.).

Soil for the SAT columns packing was obtained from the dry riverbed of the Songhua River (Harbin, China) and further sieved to guarantee the diameter of soil particles being less than 2 mm. The packed soil samples had an average pH value of 8.1, organic carbon content (OC) of 3.1%, and cation exchange capacity (CEC) of 7.5 $cmol/kg$. The results of the particle size measurement indicated that the composition of the soil was 48.5% sand, 45.1% silt, and 6.4% clay, respectively. A XRF analysis indicated that the soil samples contained SiO_2 (61.5%), Al_2O_3 (16.4%), Fe_2O_3 (4.8%), K_2O (2.8%), MgO (1.7%), Na_2O (1.5%), CaO (1.3%), TiO_2 (0.7%), and LOI (8.2%).

Secondary effluent was obtained from the Taiping WWTP, Harbin (China), which has a design capacity of $3.25 \times 10^5 m^3/d$. Collection of secondary effluent was performed at least once a month between February 2011 and August 2012. The secondary effluent had a pH value of 7.8 ± 0.1 , chemical oxygen demand (COD) of $13.7 \pm 2.3 mg/L$, DOC of $11.4 \pm 1.5 mg/L$, ultraviolet light adsorption at 254 nm (UV-254) of $19.3 \pm 2.4 m^{-1}$, suspended solids (SS) of $11.2 \pm 6.2 mg/L$, ammonia of $7.7 \pm 1.9 mg N/L$, and total phosphorus of $3.4 \pm 1.3 mg/L$.

2.2. Equipment and methods

XRD patterns of the experimental coal slag were collected with a Rigaku 2400 diffractometer (DMAX-2400, Rigaku, Japan) using Cu $K\alpha$ ($\lambda = 1.5406 \text{ \AA}$) at step scan of 0.02° from 10° to 80° . The applied current and accelerating voltage were 20 mA and 40 kV. BET specific surface area of the coal slag was obtained using a Shimadzu 2360 micrometrics automatic surface area analyzer (Japan). The pH of water solutions and soil samples was measured using a PHS-3TC pH meter (Rex, Shanghai Precision Scientific Instrument Co., China). Electrical conductivity (EC) was determined from suspension with a 1:1 soil: water ratio according to USEPA Method 9050 using conductivity meter (DDS-11A, Shanghai Hongyi Instrument Factory). CEC was measured according to the USEPA Method 9081, by mixing soil with excess sodium acetate solution, and the concentration of displaced sodium was determined by atomic absorption emission spectroscopy.

DOC, UV-254 and EEM spectra of the DOM and its fractions were measured by a TOC-5000 Total Organic Carbon Analyzer (Shimadzu, Japan), a UV-2550 UV/VIS spectrophotometer (Shimadzu, Japan) and a Jasco FP-6500 spectrofluorometer (Tokyo, Japan), respectively. Specifically, each water sample was diluted to 1 mg/L of DOC with 0.01 mol/L KCl before EEM measurement, and was further acidified to pH 3 with HCl. The EEM spectra were obtained by scanning the sample over excitation wavelengths from 220 to 400 nm with 5 nm steps and emission wavelengths from 280 to 480 nm with 1 nm steps. A detailed description of these detection methods can be found in our previous studies (Xue et al., 2008; Wei et al., 2009b).

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